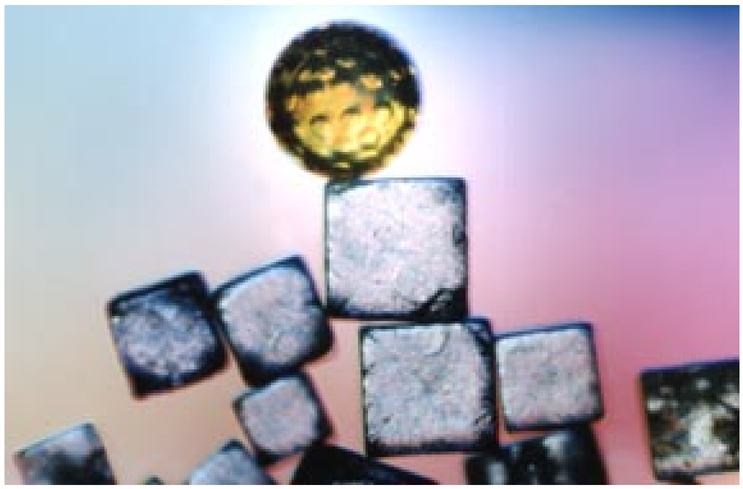
Creating Microsphere Targets for Inertial Confinement Fusion Experiments



At the heart of inertial confinement fusion (ICF) experiments are tiny, hollow microspheres used as targets. Their size, materials of construction, production methods, and structural characteristics have evolved to meet the demands of doing experiments on high-powered lasers such as Nova.

WHEN heavy isotopes of hydrogen are fused to create energy, the nuclei of the isotopes must be confined as a plasma for a period of time, depending on the density and temperature of the plasma. In a star, this confinement is accomplished by the force of the star's gravity. In the laboratory, two approaches are being pursued currently. In magnetic fusion, strong magnetic fields are used to contain relatively low-density plasmas for a few seconds. In inertial confinement fusion (ICF), the confinement times are no longer than 100 picoseconds (1 ps = 1 trillionth of a second), but the plasma densities achieved can be greater than the density of lead.

At Lawrence Livermore National Laboratory, these high-plasma densities are achieved by symmetrically depositing several kilojoules of energy from the powerful Nova laser system on a small plastic capsule containing the gaseous isotopes of hydrogen—deuterium (D_2) or deuterium–tritium (DT)—over a period of a nanosecond (1 ns = 1 billionth of a second). The deposited energy ablates the capsule wall, and the rocketlike blowoff of the hot surface material compresses the interior fuel. Although the laser system that provides the energy is as large as a football field, the target containing the fuel is only about a half millimeter in diameter. The success of each ICF experiment using this large, complex machine depends greatly on the structure and our characterization of these tiny targets. Their size, structure, and materials of construction must be perfectly suited to the particular ICF experiment for which they are used so that the resulting data can be appropriately interpreted. For more than 20 years, therefore, research concerning the design, structure, production, and performance of these capsules has been an integral part of the Laboratory's Laser Program.

ICF Target History

Targets for the first ICF experiments at LLNL were glass shells produced from an aqueous glass solution. Small droplets of this solution were dried in a heated drop tower to form hollow shells. These glass targets, 70 micrometers (μm) in diameter, offered the advantages of high strength, low gas permeability, easy doping with diagnostic atoms, and excellent symmetry and surface finish. As the laser drivers improved, larger and higher quality glass shells were fabricated. However, the high density and atomic mass of glass limited the variety of possible experiments, and so an alternative material with a lower atomic mass was sought.

Polyvinyl alcohol (PVA) shells appeared to be an excellent alternative to glass. These shells were also prepared using heated drop-tower technology. Droplets of PVA solution about 200 μm in diameter with internal argon bubbles 100 μm in diameter were generated using a dual-orifice system. In the heated drop-tower column, these hollow droplets dry to form high-quality shells with diameters up to about 250 μm. PVA, unlike most other polymers, is particularly well suited as a capsule material because it is an outstanding diffusion barrier to the hydrogen isotopes that fill the targets to fuel ICF experiments. However, attempts to extend the size range of PVA shells to meet the needs of Nova experiments were unsuccessful, and so yet another material was sought.

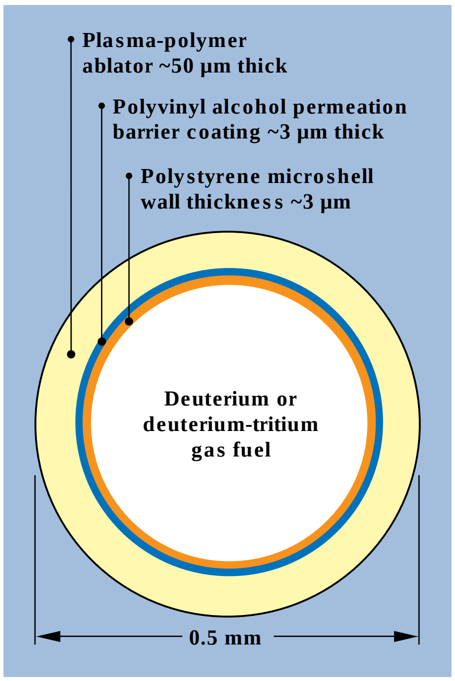


Figure 1. A typical ICF capsule, which is built around a thin polystyrene microshell. This microshell is first coated with a thin polyvinyl alcohol (PVA) layer, which serves as a permeation barrier to the gaseous fuel fill. A relatively thick ablator layer is applied using plasma polymerization techniques. These composite capsules are then filled with a precise amount of deuterium or deuterium–tritium by placing the capsule in contact with the fill gas at high pressure and elevated temperature.

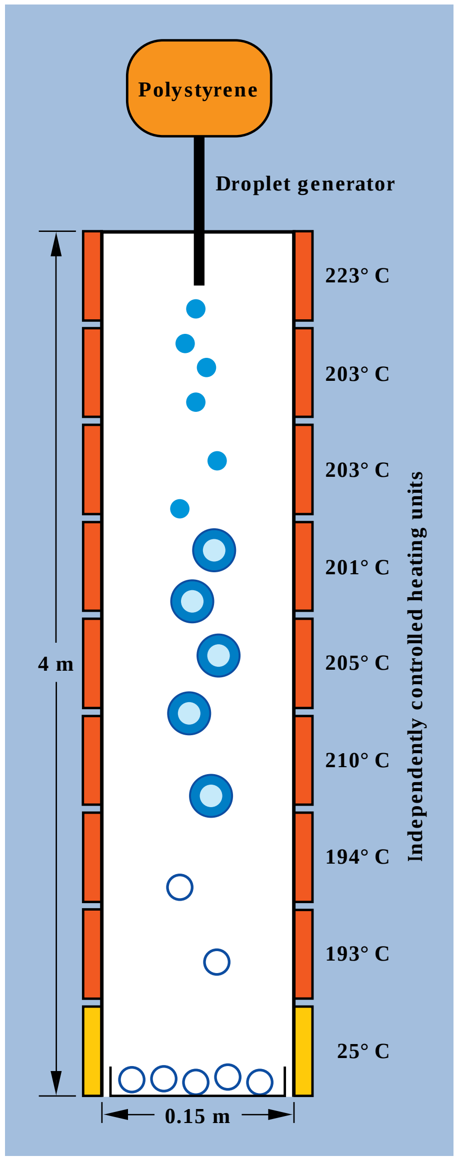


Figure 2. Diagram of the heated drop tower (not to scale) used to produce the microshells. Droplets of polystyrene (PS) solution 400 μm in diameter fall down a 4-m-high column whose temperature profile is controlled by nine independent heating units. Several thousand shells 400 to 500 μm in diameter can be produced in just a few minutes.

Current Target Capsules

The current ICF target capsule is built around a roughly one-half-millimeter-diameter polystyrene (PS) microshell and is composed of three layers (Figure 1). The innermost PS microshell is produced by the solution drop-tower technique illustrated in Figure 2. The process begins with a 4.5 wt% solution of monodisperse PS (molecular weight = 98,500) in dichloromethane that contains 3.0 wt% 2-propanol. The droplet generator provides uniform drops with diameters of about 400 μm. The drops fall through a 4-m-high tower composed of nine individually heated subunits. The first eight are heated to about 200°C; the bottom unit is kept at room temperature to cool the shells. Several thousand 400- to 500-μm-diameter shells are typically made in a run lasting a few minutes.

The shell formation process is shown in Figure 3. Initially, as the PS solution droplets fall down the tower, they shrink due to evaporation of the volatile dichloromethane solvent. As the solvent evaporates, the concentration of polymer grows near the drop surface. During this period, the drop is being heated by the surrounding gas but is cooled by evaporation. As a polymer membrane forms at the surface (accelerated somewhat by the less volatile 2-propanol, which does not dissolve PS), evaporation of the solvent (and thus cooling) is inhibited and the droplet begins to heat. When the droplet temperature exceeds its boiling point, an internal vapor bubble forms, and the shell inflates quickly. During shell expansion, evaporation is enhanced because the polymer-rich layer is thinned, allowing for better solvent transport to the surface, while the surface area is increased. The final size and quality will depend on the cooling rate and symmetry of the hot PS shell.

It is also possible to use these techniques to prepare microshells doped with small concentrations of atoms with high atomic numbers that are used as diagnostics in various ICF implosion experiments. These doped microshells are prepared from polystyrene or polystyrenelike copolymers in which the desired diagnostic atom has been bound to the polymer chain. Typical dopants are chlorine, bromine, iodine, iron, chromium, and, most recently, titanium.

The PS microshell is then coated with 2 to 3 μm of PVA, the second layer of the target capsule. This layer is necessary because PS is a poor barrier to hydrogen diffusion, and the hydrogen-isotope fuel that is eventually put in the target would leak out before it could be used. The PVA layer is applied by collecting several hundred PS microshells in a capillary tube, drawing a 10% aqueous PVA solution up around them, and then expelling them into a second heated drop tower. The solution around the individual shells dries as they fall down the tower. The efficiency of this step is low: typically the number of "target-quality" shells recovered is less than 5% of the total run. Shell loss is due largely to uneven coatings and particulate adhering to the PVA coatings.

The third and outermost capsule layer, called the ablator, is typically 30 to 60 μm thick and is deposited using plasma polymerization coating techniques. A mixture of hydrogen and *t*-2-butene at a combined pressure of about 70 mTorr is fed through a glass tube wound with wire connected to a 40-MHz radio-frequency (rf) power source. This rf power creates a plasma in the tube that breaks up the organic feed gas into molecular fragments that rain down onto the PVA-coated microshells that are agitated in a bounce pan below. The coatings produced are heavily crosslinked polymeric material and are applied at rates from 0.3 to 1.0 μm/h, depending upon the feed gas pressure and power. For some target designs, we dope the ablator with 1 to 2% germanium atoms to modify the energy absorbing properties of the coating during the implosion. This doping is done by adding a small amount of tetramethyl germane to the feed gas flow. The plasma–polymerization coating process produces a very smooth coating with a peak-to-valley variation of 100 Å (1 angstrom = 1 ten-billionth of a meter, or 10^{-10} meter) or less.

Capsule Performance

In an ideal implosion experiment, a perfectly uniform, intense energy flux would bathe a perfectly smooth and spherical capsule, ablating the outer plastic coating and compressing the fuel inside. In this situation, the performance of the capsule, generally measured as neutron yield, would depend only upon the power delivered to the capsule surface. However, in real experiments, capsule performance is degraded due to both spatial non-uniformities in the energy flux and

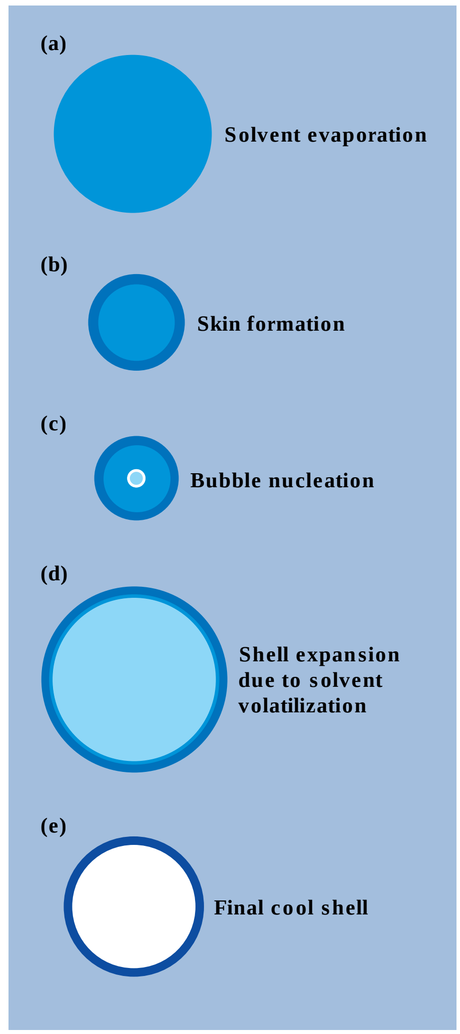


Figure 3. The shell formation process begins when the solvent rapidly evaporates from the polymer solution droplet (a) and a skin forms at the droplet surface (b). The skin retards evaporation, and the droplet begins to heat. When the droplet temperature exceeds its boiling point, a vapor bubble forms inside the shell (c) and inflates rapidly (d). The final size and quality of the shell (e) depend on the cooling rate and the symmetry of the hot PS shell.

capsule imperfections. In very simple terms, the basic principle is that the smoother and more symmetric the energy flux or capsule, the more even and efficient the implosion. Conversely, the more uneven the energy flux or rougher the capsule surface, the greater the target performance degradation, because hydrodynamic instabilities in the capsule during the ablation phase of the implosion lead to nonuniform and inefficient compression.

Because it is impossible to produce a capsule that is perfectly smooth and perfectly spherical, we are working to verify our understanding of the effects of capsule imperfections on ICF implosions through current Nova experiments. This verification is important to strengthen our case that we can produce a capsule that will ignite under the conditions planned for the National Ignition Facility (NIF). From the capsule point of view, these experiments have required us to develop the capability of making very precise measurements of what the capsule surface irregularities actually are. These measurements, which we call surface maps, provide the input data for the theories that predict capsule performance and allow us to compare experimental results with theoretical understanding.

Before we discuss how we measure capsule surface roughness and use this information to predict capsule performance, we need to understand the nature of the hydrodynamic instability mentioned above. Technically it is called Rayleigh–Taylor instability, after Lord Rayleigh, who investigated it more than 100 years ago, and Taylor, who investigated it experimentally in the 1950s. The basic phenomenon is simple (Figure 4). Imagine a situation in which we very carefully create a layer of dense fluid on top of a lighter, less dense fluid. For the purposes of this example, think of the light fluid uniformly pushing against the dense fluid and holding it up. As long as the interface between the fluids is perfectly flat and horizontal, the fluids can remain in place. However, if a small perturbation is created at the interface, the system becomes unstable and the amplitude of the perturbation will grow, allowing the more dense fluid to flow down as the lighter fluid pushes up through it.

An analogous situation exists during the ablation phase of an ICF implosion. In this case, the less dense plasma created during ablation is pushing against the dense capsule surface. Perturbations on the surface grow during this process (Figure 5). That growth can lead to a degradation of the capsule performance because compression efficiency is lost and the growing perturbations cause the inner capsule surface to mix with the fuel and cool it.

The degree of growth of perturbations on a capsule surface depends upon their "mode number." To understand this concept, suppose we trace Earth's surface, starting at the north pole, passing through the south pole, and returning to the north pole. This trace looks like a circle. A more careful measurement, however, shows that the diameter of this circle is a little less if measured between the poles than if measured at the equator. This type of asymmetry is called a "mode 2" feature, because it has two cycles per circumference.

ICF capsules also generally have a mode 2 asymmetry that can have an amplitude of a few micrometers, or about 1% of the capsule radius. Although this is by far the largest amplitude asymmetry in a capsule, it is relatively stable during the

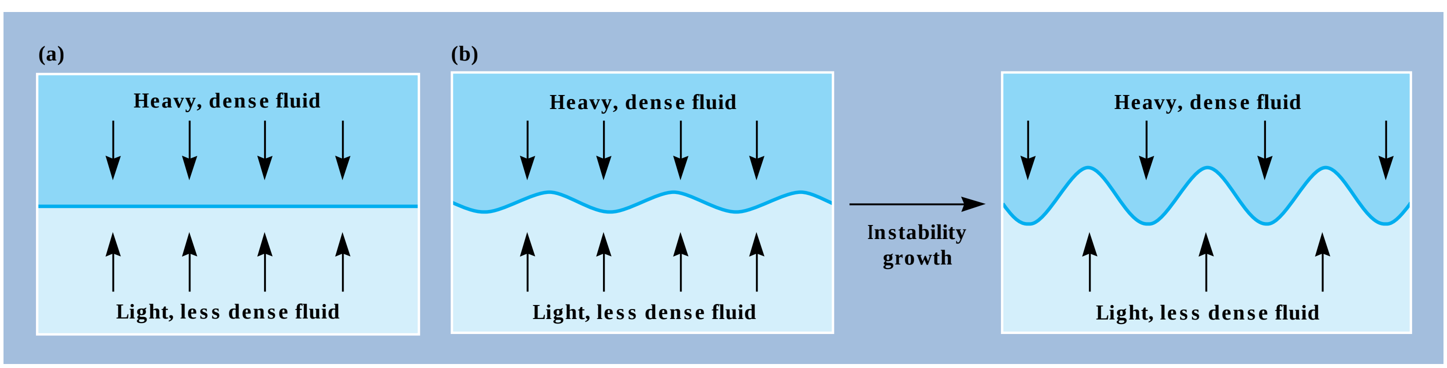


Figure 4. Illustration of the concept of the Rayleigh–Taylor instability that degrades ICF capsule performance. (a) As long as the interface between a heavy, dense fluid and a light, less dense fluid is perfectly flat and horizontal, the fluids can remain in place. (b) However, if a small perturbation is created at the interface, the system becomes unstable and the amplitude of the perturbation will grow, allowing the more dense fluid to flow down as the lighter fluid pushes up through it. In an ICF implosion, low-density ablated plasma is pushing against the dense capsule surface; thus surface perturbations at the capsule surface are Rayleigh–Taylor unstable and grow as illustrated in Figure 5.

implosion and does not grow appreciably.

Consider again our trace of Earth. If we look very carefully over very short distances along the trace, we see small bumps with amplitudes up to a few hundred feet due to trees and houses, and over even shorter distances, smaller bumps due to people, stones, and blades of grass. This manifestation of surface roughness in the Earth trace represents extremely high mode numbers but relatively low amplitudes. High mode defects of this kind are also present on ICF capsules, but they also do not grow appreciably during the compression. Intermediate between the very low mode features and the high mode features are modal features between, say, mode 10 and 100 (at length scales between one-tenth and one-hundredth of the trace circumference). In our example of the Earth trace, these are features like continents or mountain ranges on continents. The amplitudes of these features would be from one to several miles (approximately 0.025% to 0.1% of Earth's radius), much less than the amplitude of Earth's mode 2 asymmetry and much greater than he amplitudes of the higher mode features.

ICF capsules also have surface roughness in this modal range, at lateral length scales between a few tens to a few hundreds of micrometers and amplitudes up to approximately 0.025% of the capsule radius. It turns out that the amplitudes of these modes grow the most during the implosion, and thus control of surface roughness over these modes is extremely important.

Mapping the Capsule Surface

To understand, and thus predict, the effects of capsule surface roughness on performance, we have had to develop ways of mapping capsule surface roughness, particularly in those modal regions that lead to the maximum growth during the implosion.

Because of the sensitivity of the Rayleigh–Taylor instability to mode number, it is important to be able to characterize the capsule surface asymmetries over lateral length scales up to hundreds of micrometers with a vertical resolution of about 10 Å. At LLNL, we have developed a measuring device called a "Sphere Mapper," based on an atomic-force microscope (AFM) (Figure 6). The capsule is supported on a vacuum chuck connected to an air-bearing rotor. Its equator is positioned next to a stand-alone AFM head, and the

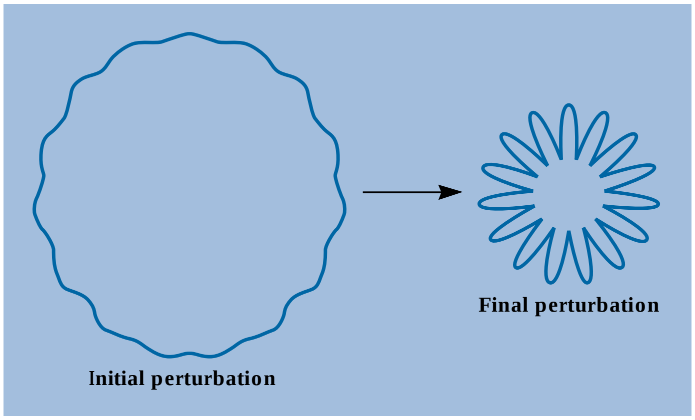


Figure 5. During an implosion, small amplitude perturbations on the capsule surface can grow by factors of 100 or more, depending upon the specific details of the perturbation and the implosion. This can lead to a degradation of capsule performance.

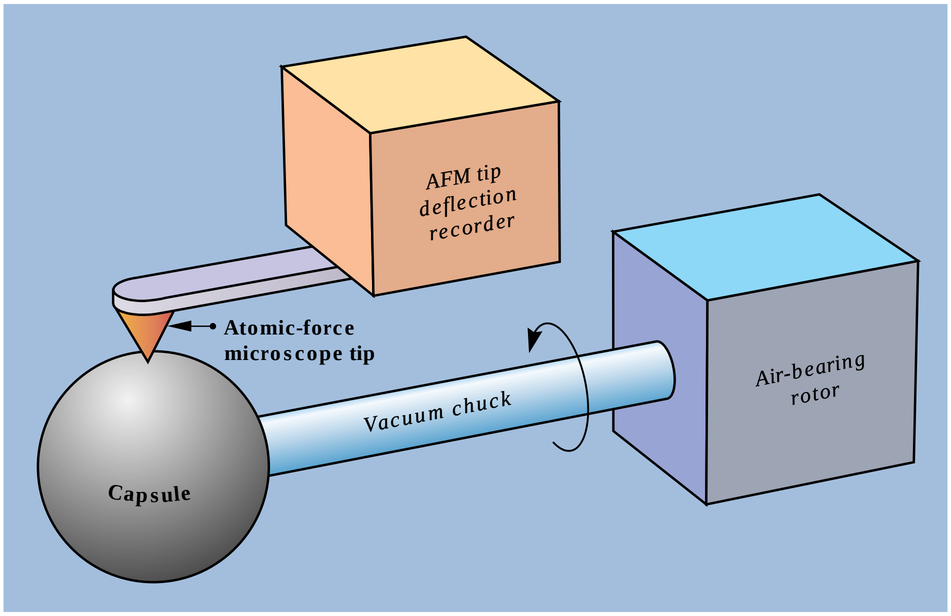


Figure 6. Schematic diagram (not to scale) of the Sphere Mapper, which is used to characterize the capsule surface finish. As the capsule is rotated, an atomic force microscope (AFM) records variations in the capsule radius to a precision of about 10 Å. Typically, three orthogonal sets of three traces are taken, as shown in Figure 7.

capsule is rotated while the AFM records height measurements at 3600 evenly spaced points. Generally three sets of equatorial traces are taken: the Sphere Mapper takes traces at the equator and at 20 μm above and below it; then the capsule is rotated 90 degrees and the process repeated; one additional 90-degree rotation gives us three orthogonal sets of three traces, as shown in Figure 7.

For comparison, typical trace data for a 1000-μm-diameter precision ball bearing made of silicon nitride and for a 435-μm-diameter titanium-doped microshell are shown in Figure 8. Note that the vertical scale is in tenths of micrometers while the horizontal scale is in degrees of rotation. The translational distance per degree is shown as an inset in each plot. Thus, the apparently sharp spikes in Figure 8a are in reality several micrometers wide. In the data in Figure 8b, the small bump at about 160 degrees is about 0.025 μm high and 60 μm wide.

Note that the ball bearing is extremely spherical but relatively rough on a short-length scale. In contrast, the capsule shows a significant (by comparison) mode 2 asymmetry but is locally extremely smooth. Also notable on this capsule are some intermediate-length scale perturbations, 20 μm to approximately 100 μm in breadth, with amplitudes of 0.01 to 0.1 μm. These are the kind of surface perturbations associated with large Rayleigh–Taylor instability growth, i.e., degradation of implosion performance,

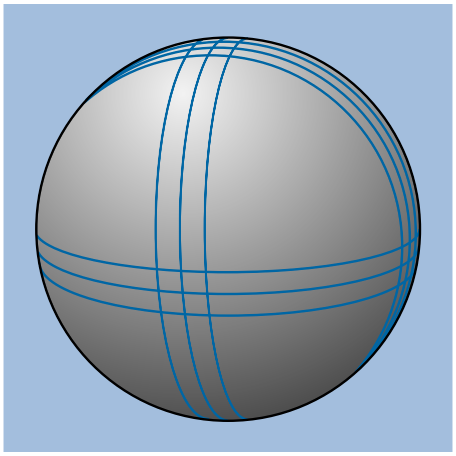


Figure 7. The Sphere Mapper takes three orthogonal sets of three traces to characterize the surface finish of ICF capsules. Example trace data are shown in Figure 8.

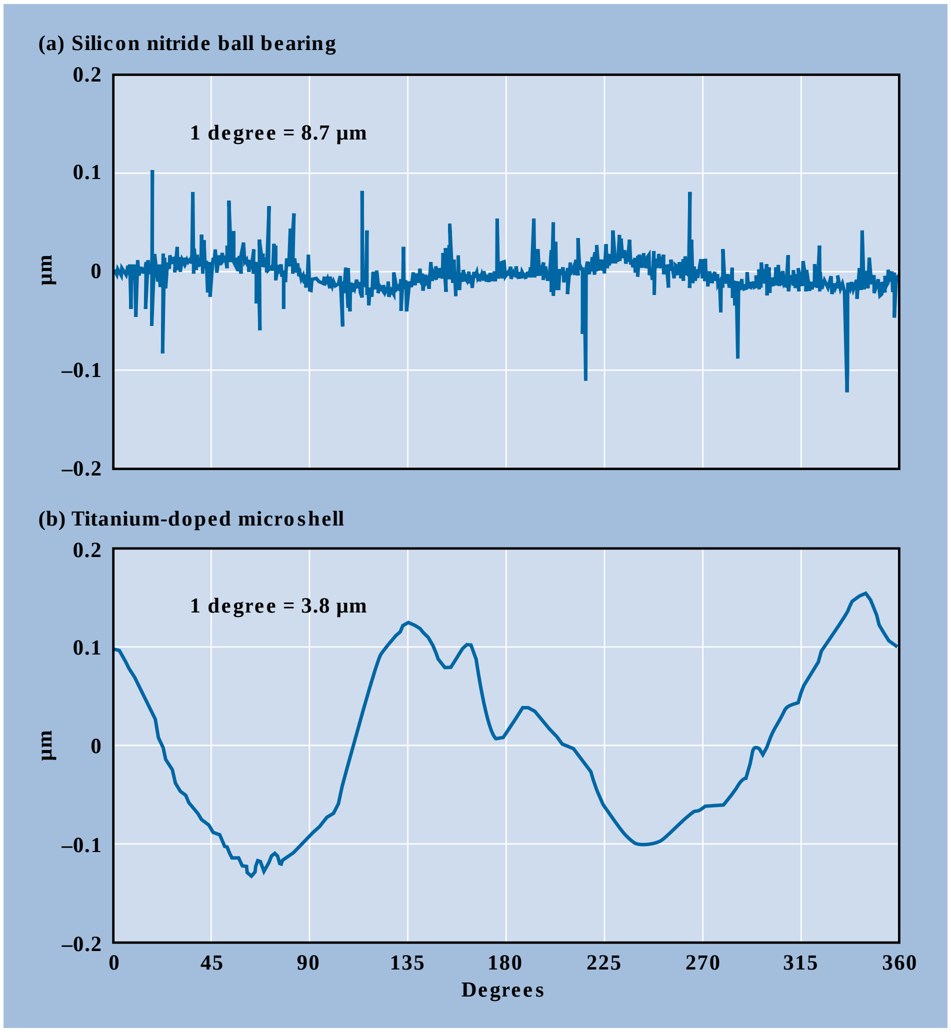


Figure 8. Typical trace data from (a) a silicon nitride ball bearing and (b) a titanium-doped microshell. Note that the horizontal axis is in degrees of rotation. The translational distance along the capsule surface associated with 1 degree of rotation is given on each plot and is much greater than the displayed vertical scale. The relatively shallow, intermediate-length-scale perturbations of the microshell surface are associated with the largest Rayleigh–Taylor growth during an ICF implosion.

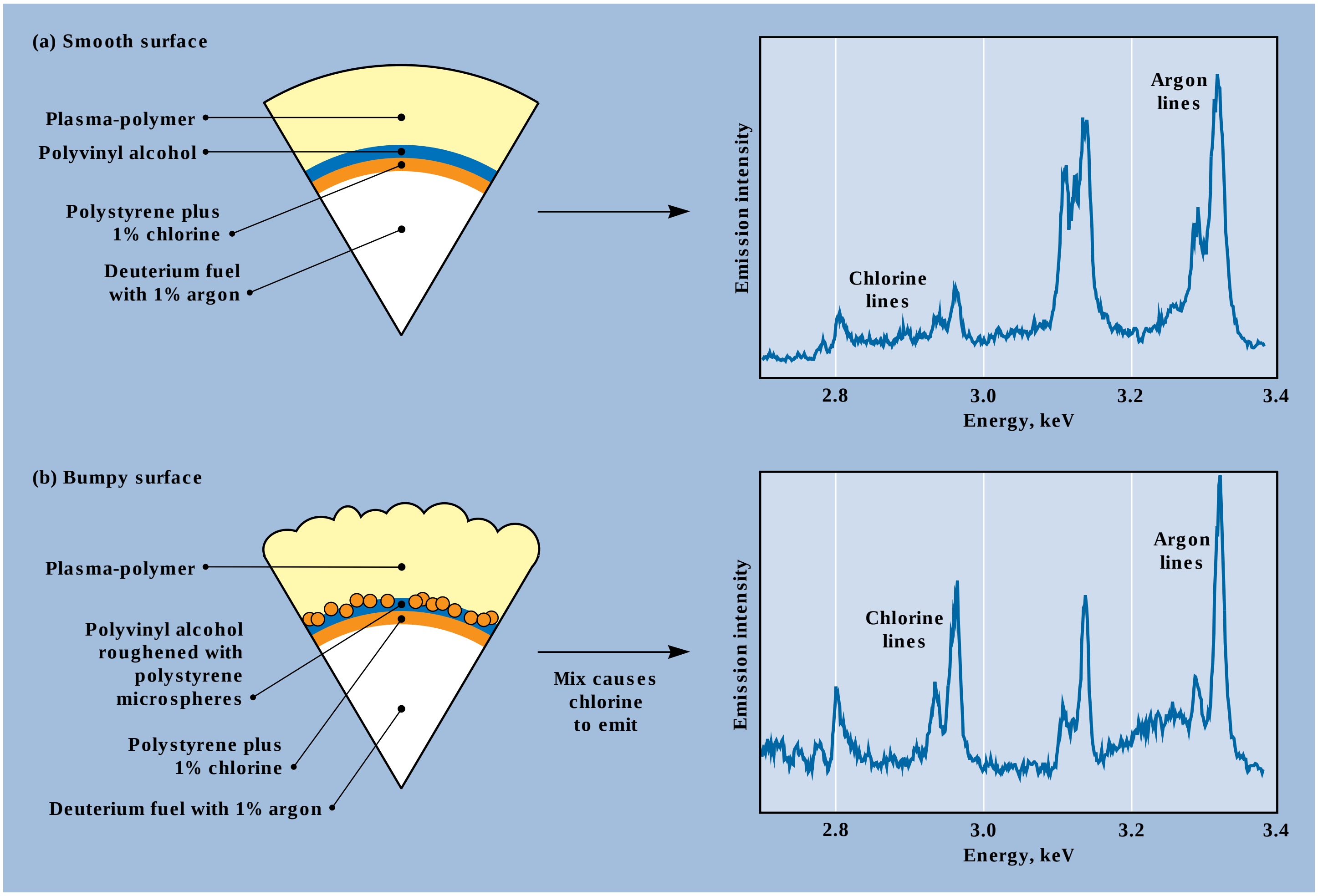
while the mode 2 asymmetry with an amplitude of about 0.3-μm has little effect on an ICF implosion.

Surface Modification

To determine the effects of capsule surface finish on capsule performance, we developed controlled ways of varying the outer capsule topography. Initially we made use of the fact that the plasma–polymer coating is conformal, and that deliberate perturbations on the surface of the PVA layer would manifest themselves in perturbations on the outside of the finished capsule. Our approach to roughening the PVA layer prior to plasma polymer coating was to include 2- to 4-μm-diameter solid polystyrene microspheres in the PVA coating solution. These microspheres were thus incorporated into the PVA coating, producing a bumpy surface on the completed capsule as shown in the sketch at the left in Figure 9b.

In these experiments, incorporation of dopants in the polystyrene

Figure 9. **On the left are sketches of (a) a smooth and (b) a bumpy capsule. On the right are the emission spectra obtained during the implosion of these capsules. The bumpy capsule (b) leads to more mix and thus a more intense chlorine signal than does the smooth surface (a). Data from experiments with modified targets such as this enable us to validate theoretical models of target performance.**



microshell and the gaseous fuel allowed spectroscopic diagnosis of the degree of "mix" of the inner capsule wall with the fuel due to the growth of surface perturbations. (Compare Figure 9a and 9b.) For a smooth capsule (Figure 9a), there was little mix, and the inner wall was kept well outside of the hot central fuel region, resulting in only a small chlorine emission in comparison to the emission from the argon dopant in the fuel. For the rough capsule (Figure 9b), however, the growth of surface perturbations resulted in a much greater chlorine emission signal when compared with the signal from the fuel argon dopant, indicating a significant mixing of the inner capsule wall with the hot fuel.

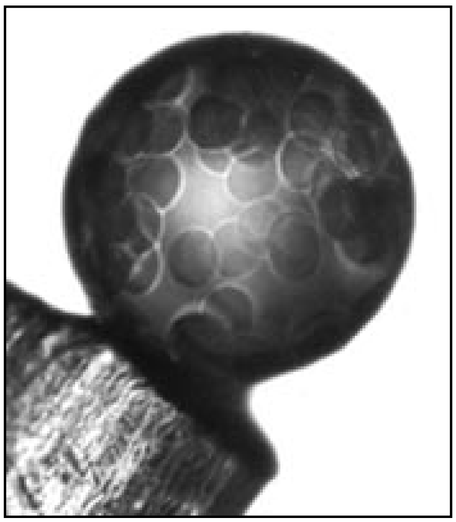


Figure 10. **A 0.5-mm-diameter plasma–polymer-coated capsule with 200 randomly placed ablated pits, each about 75 μm in diameter and 1 μm deep. This capsule was intentionally roughened to a predetermined extent to study how surface perturbations affect capsule performance during an implosion.**

The use of polystyrene seeds in the PVA layer as a mechanism to produce roughened capsule surfaces offered only modest control over the degree of roughness. Recently we have developed the capability of using a laser to precisely carve pits with depths from 0.1 to 3 μm and widths up to 100 μm on capsule surfaces. In Figure 10, the photo shows a 0.5-mm-diameter plasma–polymer-coated capsule with 200 randomly placed pits, each roughly 75 μm in diameter and 1 μm deep. The cover photo shows a capsule with symmetrically placed pits. The success of this technique for roughening capsules to a predetermined extent is proving extremely valuable in developing our understanding of Rayleigh–Taylor hydrodynamic instabilities during an implosion due to capsule surface perturbations.

Future ICF Needs

As the ICF community develops more powerful laser drivers, the capsules used in these experiments will need to be larger. The Omega Upgrade ICF facility at the University of Rochester's Laboratory for Laser Energetics will come on-line in the spring of 1995 and will require capsules that are roughly 1 mm in diameter. The National Ignition Facility (NIF), with a planned completion date shortly after the year 2000, will require 2-mm-diameter capsules. The production of these capsules will depend on the development of new technology, because our solution drop-tower methods are limited to approximately 0.5-mm capsules, due largely to the large heat and mass transfers necessary.

A number of alternative technologies, however, hold promise for delivering larger capsules. The most widely developed is microencapsulation. In this method, a water droplet is encapsulated by a polymer solution and then suspended in an aqueous phase. The organic solvent containing the polymer slowly dissipates into the aqueous phase, leaving behind a polymer shell. This approach to shell manufacture has been used for a number of years at the Institute for Laser Engineering at Osaka University and also at the University of Rochester. In both cases, the typical polystyrene shells produced for ICF targets have been significantly less than 1 mm in diameter. Work is in progress to extend this technology to 1- to 2-mm capsules suitable for Omega and NIF ICF experiments.

The target fabrication group at the Lebedev Institute in Moscow has historically made plastic shells for Russian ICF experiments by heated drop-tower techniques using particles of solid polymer infused with small amounts of volatile organic solvent that inflate the particles when they are melted in the drop tower. This approach is useful for larger shells because much less mass and heat transfer is necessary compared to solution drop-tower techniques. Using this technique, the Lebedev group has been able to routinely prepare 1-mm capsules with good symmetry and surface finish. For larger shells, there is concern that the hydrodynamic interaction of the falling molten capsule with the surrounding atmosphere will lead to distortions. To remedy this problem, as well as to maximize heat transfer in the minimum drop-tower height, they have developed the concept of a "Ballistic Furnace." The polymer particles are injected up into the column so that they inflate to become hollow shells at the trajectory apex, when the hydrodynamic interactions with the surrounding media are at a minimum.

A route to larger shells as well as unique capsules for current ICF experiments is being developed at LLNL based on the use of decomposable solid or hollow mandrels. Briefly, the method makes use of the fact that poly (α-methylstyrene) thermally decomposes to a gas at a relatively low temperature. It is possible to prepare very symmetric and smooth solid beads or hollow shells of this polymer at sizes up to several millimeters, overcoat them with a layer of plasma polymer, and then heat them to decompose the poly(α-methylstyrene) mandrel, leaving a symmetric shell of the desired size. The method may also be useful for preparing capsules with prescribed *inner* surface roughness by using the laser ablation technology discussed earlier to roughen the poly(α-methylstyrene) mandrel before overcoating. In this way, the rough contours of the mandrel will be reproduced on the *inside* surface of the shell remaining after thermal treatment.

Future ICF target designs for both the Omega and NIF facilities call for cryogenic targets in which a 100-μm-thick, symmetric, solid or liquid D_2 or DT fuel layer is present on the inside of the capsule wall. One route to accomplishing this is to prepare capsules with a low-density organic foam liner to hold the fuel inside the full-density plastic shell. Capsules of this type have been developed at Osaka University and at LLNL using microencapsulation techniques. The method involves microencapsulating a droplet of water or oil by a 100-μm-thick layer of an oil or water phase, respectively, which contains about 5% polymerizable components. This layered droplet is suspended in the same phase as the inner droplet. By initiating polymerization of the dilute monomer in the layer, a solid but low-density foam structure can form. This foam shell can then be overcoated with a full-density layer and finally dried to produce the desired foam-lined capsule.

Summary

The production of ICF capsules, with their very stringent symmetry and surface finish requirements, and the characterization of them represent major materials science challenges. The quality of the capsule is largely dependent upon the quality of the thin-walled plastic microshell around which it is built. The characterization requirements for the capsules have led to the development of new and unique capabilities for measuring their surface topology. The scientific interest in studying the effects of surface topology on the implosion dynamics has led to methods of precisely modifying the capsule surface finish. Future ICF targets will require larger capsules, and technologies designed to meet this need are now being developed.

Key Words: drop-tower technology; inertial confinement fusion (ICF) target capsules; microshell; microsphere; mode growth; National Ignition Facility (NIF); Nova laser; Rayleigh–Taylor hydrodynamic instability; Sphere Mapper.

Notes and References

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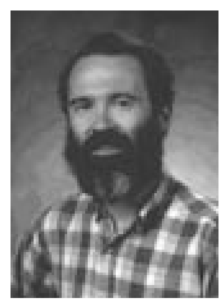

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